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Group delay dispersion measurements in the mid-infrared spectral range of 2-20 µm

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Abstract: We present two measurement devices which both allow the direct measurement of the group delay (GD) and group delay dispersion (GDD) of laser optics, covering the near- and mid-infrared (MIR) spectral range from 2 to 20 µm (500-5,000 cm^{-1}). Two different kinds of devices were developed to measure the GDD of multilayer interference coatings. One is a resonant scanning interferometer (RSI) and the other is a white light interferometer (WLI). The WLI is also capable of measuring the GDD in transmission, for instance of bulk material. GDD measurements of a high dispersive mirror for wavelengths from 2.0 to 2.15 µm and one of a multilayer mirror from 8.5 to 12.0 µm are presented. A measurement of a zinc selenide (ZnSe) substrate in transmission was made with the WLI demonstrating the full bandwidth of the device from 1.9 to 20 µm. The comparison of all measurements with their related theoretical values shows a remarkable correspondence.

OCIS codes: (120.3180) Interferometry; (320.7100) Ultrafast measurements; (310.3840) Materials and process characterization; (310.1620) Interference coatings.

References and links

1. E. R. Crosson, K. N. Ricci, B. A. Richman, F. C. Chilese, T. G. Owano, R. A. Provencal, M. W. Todd, J. Glasser, A. A. Kachanov, B. A. Paldus, T. G. Spence, and R. N. Zare, "Stable isotope ratios using cavity ring-down spectroscopy: Determination of 13C/12C for carbon dioxide in human breath," Anal. Chem. **74**(9), 2003–2007 (2002).
2. A. Schliesser, N. Picqué, and T. W. Hänsch, "Mid-infrared frequency combs," Nat. Photonics **6**(7), 440–449 (2012).
3. M. W. Todd, R. A. Provencal, T. G. Owano, B. A. Paldus, A. Kachanov, K. L. Vodopyanov, M. Hunter, S. L. Coy, J. I. Steinfeld, and J. T. Arnold, "Application of mid-infrared cavity-ringdown spectroscopy to trace explosives vapor detection using a broadly tunable (6-8 µm) optical parametric oscillator," Appl. Phys. B Lasers Opt. **75**(2-3), 367–376 (2002).
4. H. Pires, M. Baudisch, D. Sanchez, M. Hemmer, and J. Biegert, "Ultrashort pulse generation in the mid-IR," Prog. Quantum Electron. **43**, 1–30 (2015).
5. I. Pupeza, D. Sánchez, J. Zhang, N. Lilienfein, M. Seidel, N. Karpowicz, T. Paasch-Colberg, I. Znakovskaya, M. Pescher, W. Schweinberger, V. Pervak, E. Fill, O. Pronin, Z. Wei, F. Krausz, A. Apolonski, and J. Biegert, "High-power sub-two-cycle mid-infrared pulses at 100 MHz repetition rate," Nat. Photonics **9**(11), 721–724 (2015).
6. I. Pupeza, T. Eidam, J. Rauschenberger, B. Bernhardt, A. Ozawa, E. Fill, A. Apolonski, T. Udem, J. Limpert, Z. A. Alahmed, A. M. Azzeer, A. Tünnermann, T. W. Hänsch, and F. Krausz, "Power scaling of a high-repetition-rate enhancement cavity," Opt. Lett. **35**(12), 2052–2054 (2010).
7. R. Szipöcs, C. Spielmann, F. Krausz, and K. Ferencz, "Chirped multilayer coatings for broadband dispersion control in femtosecond lasers," Opt. Lett. **19**(3), 201 (1994).
8. F. X. Kärtner, N. Matuschek, T. Schibli, U. Keller, H. A. Haus, C. Heine, R. Morf, V. Scheuer, M. Tilsch, and T. Tschudi, "Design and fabrication of double-chirped mirrors," Opt. Lett. **22**(11), 831–833 (1997).
9. V. Pervak, O. Razskazovskaya, I. B. Angelov, K. L. Vodopyanov, and M. Trubetskov, "Dispersive mirror technology for ultrafast lasers in the range 220–4500 nm," Adv. Opt. Technol. **3**, 55–63 (2014).
10. T. Brabec and F. Krausz, "Intense few-cycle laser fields: Frontiers of nonlinear optics," Rev. Mod. Phys. **72**(2), 545–591 (2000).
11. T. V. Amotchkina, A. V. Tikhonravov, M. K. Trubetskov, D. Grupe, A. Apolonski, and V. Pervak, "Measurement of group delay of dispersive mirrors with white-light interferometer," Appl. Opt. **48**(5), 949–956 (2009).

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12. M. K. Trubetskov, M. von Pechmann, I. B. Angelov, K. L. Vodopyanov, F. Krausz, and V. Pervak, "Measurements of the group delay and the group delay dispersion with resonance scanning interferometer," Opt. Express **21**(6), 6658–6669 (2013).
13. A. P. Kovács, K. Osvay, Z. Bor, and R. Szipöcs, "Group-delay measurement on laser mirrors by spectrally resolved white-light interferometry," Opt. Lett. **20**(7), 788–790 (1995).
14. W. H. Knox, N. M. Pearson, K. D. Li, and C. A. Hirlimann, "Interferometric measurements of femtosecond group delay in optical components," Opt. Lett. **13**(7), 574 (1988).
15. W. H. Knox, T. B. Laboratories, and C. C. Road, "Dispersion measurements for femtosecond-pulse generation and applications," Appl. Phys. B **235**, 225–235 (1994).
16. K. Osvay, G. Kurdi, J. Hebling, A. P. Kovacs, Z. Bor, and R. Szipocs, "Measurement of the group delay of laser mirrors by a Fabry-Perot interferometer," Opt. Lett. **20**(22), 2339 (1995).
17. K. Naganuma, K. Mogi, and H. Yamada, "Group-delay measurement using the Fourier transform of an interferometric cross correlation generated by white light," Opt. Lett. **15**(7), 393–395 (1990).
18. S. Diddams and J.-C. Diels, "Dispersion measurements with white-light interferometry," J. Opt. Soc. Am. B **13**(6), 1120 (1996).
19. J. Connolly, B. DiBenedetto, and R. Donadio, "Specifications Of Raytran Material," in *Techincal Symposium East*, R. E. Fischer, ed. (International Society for Optics and Photonics, 1979), pp. 141–144.

1. Introduction

In the 'finger print' region between 2 and 20 μm, most molecules have fundamental vibrational modes, making mid-infrared (MIR) spectroscopy an important tool for many fields including quality control, food industry, forensic analysis, semiconductor electronics, biomedical applications and many others [1–4]. To increase the signal-to-noise ratio in optical spectroscopy, spatial coherent and high power light is desired. Recently a source of coherent radiation with a spectrum spanning the range from 6.8 to 16.4 μm and an average power of 0.1 W was developed [5]. This phase coherent source with a repetition rate of 100 MHz enables the resonant enhancement of radiation in a passive cavity and thus increases the interaction length with the examined medium. The phase of a pulse circulating in such an enhancement cavity must be well controlled by dispersive mirrors [6]. The development and production of multilayer optics for MIR applications require metrological support providing a valuable feedback information and quality verification. In particular the measurement of the group delay (GD) and group delay dispersion (GDD) in the MIR is essential for the development of dispersive (or chirped) mirrors [7–9] for the MIR spectral range, since these characteristics determine the shape of pulse envelopes [10].

Recently GDD measurements in the UV-vis-NIR optical region using a home-made WLI [11] and a RSI [12] were published. In this work, we use the same principals as mentioned in these publications. Unfortunately, none of the optical components of the existing setups are suitable for MIR radiation. In the previous devices the spectrometer, light source, beam splitter and partial reflectors were selected for a spectral region up to only 2 μm. In the MIR wavelength range typically Fourier transform infrared spectrometers (FTIR) are used, but they are less sensitive than VIS-NIR spectrometers. As a result the MIR interferograms measured with the WLI are more noisy. That makes it impossible to use the old evaluation algorithms. Extending the previously known working principles to the MIR wavelength region required the search for new optical components, light source, spectrometer and a new evaluation software, resulting in the development of completely new devices.

2. Measurement setups

All optical components, the light source and the spectrometer, must be designed to work in the MIR spectral range. The dominating type of spectrometers for the MIR are typically FTIR spectrometers. Besides also dispersive grating spectral photometers exist, but they are too insensitive for our applications. We use a FTIR spectrometer Vertex 70 from Bruker Opik GmbH, Germany. The device is a stand-alone FTIR spectrophotometer which allows transmittance and reflectance measurements of optical components, but it also has an external input to measure the light from our interferometers described below. For the high acquisition rates needed for the WLI, the FTIR spectrometer is equiped with a sensitive mercury

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cadmium telluride (MCT) detector, which is cooled with liquid nitrogen and has a working range of 1.25 to 20 µm (8000 to 500 cm^{-1}). For the incoherent MIR light source we used a silicon carbide globar (Newport Corp.) which emits radiation from 1.7 to 25 µm. Most of the optical components were coated in-house with an electron beam deposition system described in section 3.2. The alignment of a MIR-optical path was found to be very challenging, since we couldn't find a way to make the MIR light visible. Therefore, we produced gold mirrors which have some residual transmittance of about 1% in the visible range. Due to this one can see the glowing filament of the globar by eye, looking through the backside of the gold mirrors. This method was found to be most practical for aligning the optical setups.

Two different approaches to measure the GDD of optics were realized. The first approach is based on a resonant scanning interferometer (RSI) [12]. The second measurement setup is a scanning white light interferometer [11,13–15].

2.1 Resonant scanning interferometer (RSI)

An RSI is basically a Fabry-Perot interferometer consisting of two semitransparent mirrors and is operated with a white light source. The two mirrors are separated by a thin air spacer. The spectrum of the light transmitted by the end mirror exhibits sharp resonance peaks. From the spectral distance of the resonant peaks to each other, the group delay can be calculated when the thickness of the air spacer is known [16]. By having multiple spectrums at different thicknesses of the air spacer, the group delay can be calculated with a high spectral resolution, without knowing the distance of the two mirrors, using an advanced algorithm [12]. For our setup we build an RSI in the so-called reflection mode and measured the reflected light of the resonator (Fig. 1). Such setup of the interferometer is also called a Gires-Tournois interferometer. Measuring the reflected light is especially essential when the mirror substrate is not transparent in the spectral region of interest.

The MIR light is guided by a gold mirror to the interferometer. The partial reflector (PR) is a 3 mm thick zinc selenide (ZnSe) substrate, coated with a gold layer with a thickness of 5 nm. The sample is mounted on a translation stage to adjust the distance between the two mirrors. The reflected light with the resonance peaks is measured with the FTIR spectrometer. The interference signal only appears when the two mirror surfaces are exactly parallel to each other. For this alignment the beam of a He-Ne laser is coupled into the optical path. An interference pattern generated by the sample and the partial reflector is now observed. The tilt of the sample is carefully changed now until the interference fringes are circular. That means the two laser beams are collinear and the sample is exactly parallel to the partial reflector.

For a single GDD measurement, a few spectra are acquired at different thickness of the air-spacer. 10 spectra are typically enough to get a reliable result. The spectra are evaluated using the algorithm recently developed by Trubetskov et al. [12].

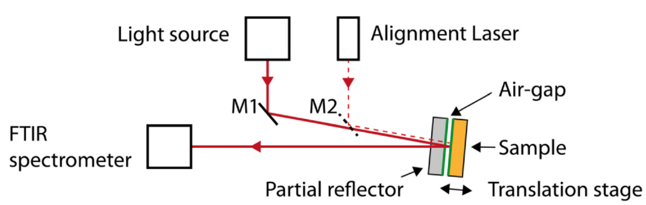

Fig. 1. Optical setup of the resonant scanning interferometer (RSI): Light is guided under a small angle into the Gires-Tournois interferometer. The distance between the partial reflector and the sample can be changed by the translation stage. The alignment laser is needed to adjust the sample to be parallel to the partial reflector

2.2 White light interferometer (WLI)

A WLI is a Michelson interferometer with a broadband incoherent light source. When both arms of the interferometer have the exact same length, interference fringes are visible at the output, where the two beams overlap. By increasing and decreasing the path length of one arm, the interference vanishes because of the short coherence length of the light source. Different methods and algorithms exist to measure and calculate the group delay from WLI measurements [13,14,17,18]. In our setup the light at the output is measured with a spectrometer. The signal for each wavelength of the spectrometer versus the path length is an interferogram. For each wavelength an envelope of the interferogram is calculated and the position of the maximum in respect to the delay is determined. For the case of an empty WLI with no dispersion, that's when the end mirrors are metal mirrors, the maxima of the interferograms appear all at the same path length. When one end mirror is exchanged for a dispersive mirror, the maxima of the interferograms can have a relative delay. That means that the penetration depth changes for different wavelength. The time delay between the maxima is exactly the group delay GD caused by the sample.

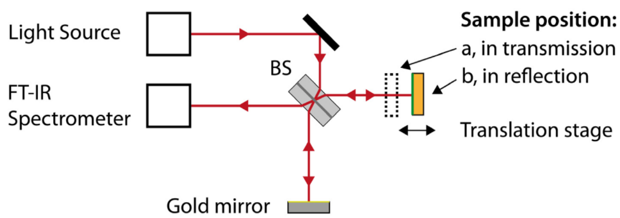

Fig. 2. Optical setup of the white light interferometer: Light is split into two arms by a thin film beam splitter. The compensation plate is directly put onto the beam splitter. The end mirrors are gold mirrors. If a sample is measured in reflection, it replaces a gold mirror and is used as an end mirror. For transmission measurements the sample is placed in the optical path of one arm.

The WLI setup is shown in Fig. 2. The light source and the FTIR spectrometer are the same as used for the RSI setup discribed in section 2.1. The beam from the light source is split into two parts with a KBr thin-film beam splitter. As illustrated in Fig. 2 the compensation plate is put directly onto the beamsplitter. The compensation plate is needed in order to have the same amount of dispersion in both arms of the interferomter. One end mirror of the interferometer is mounted onto a motorized translation stage.

For a GDD measurement, the stage is put into a position out of the interference maximum, to be able to record one whole interferogram with the maximum in the middle. Then the stage starts moving and the spectrometer acquires spectra continiously. Typically 8,000 spectra with a resolution of 10 cm^{-1} are taken. Since the resolution of the FTIR spectrometer is defined in the frequency space, it depends on the wavelength. For example the resolution of 10 cm^{-1} corrosponds to a wavelength resolution of 0.1 µm at 10 µm or 25 nm at a wavelength of 2 µm. The data is evaluated with an algorithm using the Matlab programming language. The algorithm is similar to the one published by T. Amotchkina et al. [11].

3. GD and GDD measurements

In the following three sections measurements from both devices, RSI and WLI are presented. Two multilayer mirrors are measured with both devices and the data is plotted into one graph. The measurements are compared with each other and with theoretical values. At last the GDD measured with the WLI of a ZnSe substrate in transmission is presented.

3.1 Dispersive mirror for near infrared light

The coating design for the dispersive mirror was optimized to have a flat GDD from 2.0 to 2.15 µm (Fig. 3, dashed line). The dispersive mirror is produced in a reactive magnetron sputter process (Helios from Leybold Optics) using tantal pentoxide and silicon dioxide as layer materials. The measurement demonstrates that the two presented devices allow measurements in the near infrared spectral range. The measurement of the RSI and the WLI

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agree with each other in the region of interest from 2.0 to 2.15 µm and provide an average GDD value of −1,000 fs^2 (Fig. 3). Both measurements show small oscillations in this range, which are a typical indication of small layer-thickness deviations during the coating process. The thickness-errors are typically below 0.5% for our sputter process, since the coating process is very stable and well understood. The layer thicknesses are controlled by time and calibrated by an optical broadband monitor. Outside of the region of interest the WLI produced more reliable GDD data than the RSI, since the the reflectance of the mirror decreases outside the designed range and therefore the intensity and the sharpness of the resonant peaks decrease.

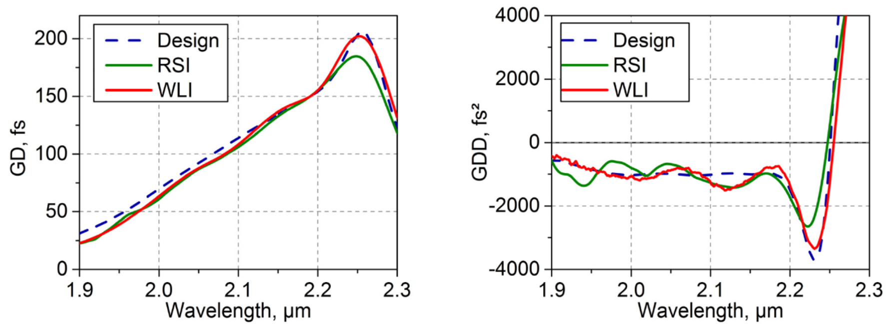

Fig. 3. High dispersive mirror designed for GDD = −1,000 fs^2 in the range of 2.0 to 2.15 µm. The oscillations are an indication for layer-thickness deviation during the coating process.

3.2 Multilayer mirror for MIR radiation

We produced a mirror for wavelengths from 8.5 to 12 µm, using an electron beam (e-beam) process using germanium (Ge) and zinc sulfide (ZnS) as layer material. The box coater is a SyrusPro from Leybold Optics, Germany. It is equiped with a quartz-crystal monitor to determine the layer thicknesses. The design is a simple quarter-wave stack (QWS) with 9 layers and a central wavelength of 10.5 µm. The first and the last layer are Ge and the coating stack is described by the formula $[Ge\ ZnS]^4\ Ge$. The substrate material is also germanium. In Fig. 4 the measurements and the theoretical values are shown.

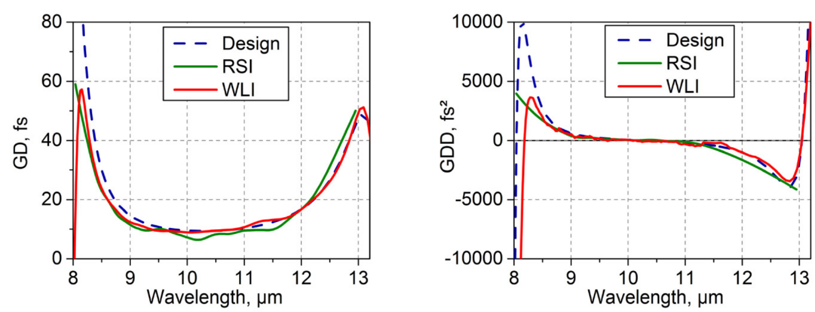

Fig. 4. Mirror for MIR light. The mirror is a QWS centered at 10.5 µm and is produced by electron beam deposition. It shows the typical shape of GD and GDD of a QWS. The characteristic is flat around the central wavelength, and the values increase when locking to shorter and longer wavelengths.

The measured values of the GD and GDD for the range of high reflectance from 8.5 to 12 µm coincide well with the theoretical values for both devices. Like in the previous section, the WLI produced more reasonable values outside of the reflectance range. We estimated the thickness errors for this e-beam coating run. Since the spectral characteristics and the GDD of a QWS are very robust versus thickness deviations, only small deviations of the GDD measurement can be seen in the region of interest between 8.5 and 12 µm. But the peak in

GDD at about 8.3 µm is obviously reduced. An error analysis of the coating design reproduced this lowered peak and indicates a random thickness-error of about 1.5%. As expected, the value is higher as for the sputter process, but it gives reason for further optimization and stabilization of our e-beam coating process.

3.3 Zinc selenide substrate

Here we demonstrate that the WLI is also capable to measure the GDD in transmission of bulk material for the whole working range from 1.9 to 20 µm. For better comparability with other substrate materials we show the group velocity dispersion (GVD), which is the GDD per unit length. We measured the GVD of a 3 mm thick Zinc selenide substrate by putting the substrate in one optical path of WLI. The end mirrors of the WLI were both gold mirrors. This kind of measurement can't be realized by RSI, because the principle of an RSI is a Fabry-Perot interferometer with a cavity length of a few ten's of micrometers.

The literature values of the wavelength dependent refractive indices n(λ) of ZnSe from 0.5 to 18.2 µm were taken from [19]. With these values the group velocity v_g and the group velocity dispersion GVD were calculated using their definitions:

$$v_g = \left(\frac{\partial k}{\partial \omega}\right)^{-1} = \frac{\partial}{\partial \omega}\left(\frac{\omega}{c}n(\omega)\right)^{-1} \text{ and } GVD = \frac{\partial}{\partial \omega}v_g^{-1} = \frac{\partial^2 k}{\partial \omega^2}$$

Here k is the wavenumber and ω is the angular frequency. The calculated values and the measurements are ploted in Fig. 5. Our measured data was taken from 1.9 to 20 µm in one single measurement scan and is in excellent agreement with the literature values.

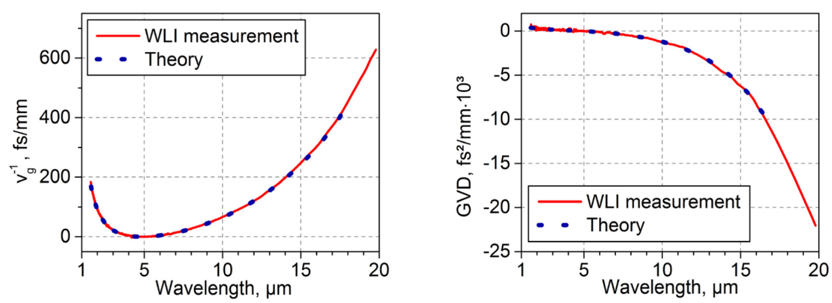

Fig. 5. Shows the inverse group velocity $1/v_g$ (left) end the GVD of ZnSe (right). The measurement is conducted using the WLI. The theoretical values are calculated from wavelength dependent refractive indices.

4. Conclusion

We have demonstrated two devices that allow the measurement of the GD and GDD of optics for ultrashort laser pulses covering the NIR and the MIR up to a wavelength of 20 µm for the first time, to our knowledge. The RSI allows fast measurements, due to a simple alignment procedure and the need of only a few spectra at different air-spacer thicknesses. The WLI produces more robust and reliable data, however the alignment of the Michelson interferometer is more challenging, and a measurement scan takes more time. GDD measurements of two interference coatings and the GVD of a ZnSe substrate show excellent agreement with theoretical values. The devices pave the way for the development of novel optical components needed for ultrashort laser sources for the MIR spectral range.

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